

Unconventional temperature dependence of the cuprate excitation spectrum

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Abstract –Key properties of the cuprates, such as the pseudogap observed above the critical temperature T_c , remain highly debated. Given their importance, we recently proposed a novel mechanism based on the Bose-like condensation of mutually interacting Cooper pairs [W. Sacks, A. Mauger, Y. Noat, Superconduct. Sci. Technol. 28 105014, (2015)]. In this work, we calculate the temperature dependent DOS using this model for different doping levels from underdoped to overdoped. In all situations, due to the presence of excited pairs, a pseudogap is found above T_c while the normal DOS is recovered at T^* , the pair formation temperature. A similar behavior is found as a function of magnetic field, crossing a vortex, where a pseudogap exists in the vortex core. We show that the precise DOS shape depends on combined pair (boson) and quasiparticle (fermion) excitations, allowing for a deeper understanding of the SC to the PG transition.

Introduction. – As is well known, the superconducting (SC) state of cuprates is characterized by a dome-shaped critical temperature T_c versus carrier density and an unconventional pseudogap (PG) state above T_c (see [1] for a review). In addition, these compounds exhibit a spatially inhomogeneous [2, 3] and unconventional quasiparticle (QP) dispersion (see [4–6] and Ref. therein), the ‘peak-dip-hump’ structure, with a characteristic gap Δ_p typically larger than the critical temperature energy scale $\sim k_B T_c$, especially in the underdoped regime.

Many of these properties remain highly debated, in particular the Cooper pair formation mechanism and the origin of the pseudogap state, which appears below the higher temperature T^* . The spectral gap is of the same order of magnitude at T_c as in the vortex core [7], indicating that the PG characterizes the loss of SC coherence [8]. In agreement with Renner et al. [9], recent experiments by Sekine et al. [10] on quasiparticle excitation spectra up to T^* are particularly intriguing. The spectral gap indeed starts to disappear and vanishes only at the higher temperature T^* .

To summarize, a solution of the high- T_c problem must account for:

- (i) multiple energy scales (T_c , T^*) and large gap Δ_p ,
- (ii) large T_c , however with $k_B T_c \ll \Delta_p$,
- (iii) the dome-shape of the $T - p$ phase diagram,
- (iv) the unconventional SC to PG transition,
- (v) the unusual QP dispersion (‘peak-dip-hump’).

With these questions in mind, we have recently proposed a model [11, 12], based on the mutual interaction between pre-formed pairs, able to take into account both phases of the system. It describes cogently the shape of the low-temperature excitation spectrum in cuprates and links the superconducting and pseudogap phases.

In an early approach, Franz and Millis [13] addressed the temperature dependence of the QP excitation spectrum based on the effects of classical phase fluctuations. Unfortunately, their model does not match the precise shape of the STS data, especially the important dip present at low temperature, a clear signature of HTc superconductivity.

More recently, Pieri et al. [14] considered pair fluctuations in the t-matrix framework, but again no dip in the zero temperature spectra was clearly demonstrated. In a related paper involving pair fluctuations [15], a repulsive

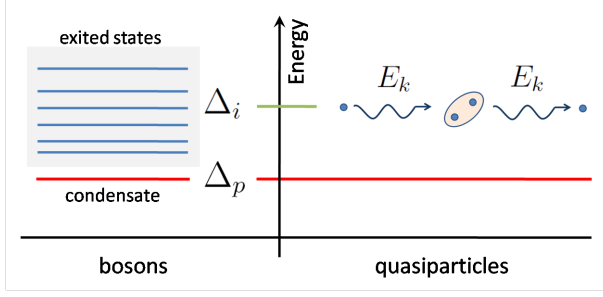


Fig. 1: Left panel: distribution of excited pair states. Right panel: energy diagram to illustrate the coupling between quasiparticles and excited pair states. Such a strong QP-pair coupling gives rise to the second term in the gap equation 2.

pair-pair interaction was inferred, like in our model, but an explicit calculation of the density of states was not reported. To the contrary, our model gives for the first time the low-temperature peak-dip-hump feature and the full temperature dependence of the QP spectra up to T^* .

At the heart of the model, an incoherent state consists of preformed pairs [16] with an energy distribution $P_0(\Delta_i)$ (see Fig. 1, left panel):

$$P_0(\Delta_i) \propto \frac{\sigma_0^2}{(\Delta_i - \Delta_0)^2 + \sigma_0^2} \quad (1)$$

where Δ_0 and σ_0 are respectively the average gap and the width of the distribution. Alternatively, $P_0(\Delta_i)$ can be considered as the density of pair states.

Pairs in the PG state have a random energy distribution, making long range order impossible. Interactions between pairs, with characteristic coupling energy β_k , allows them to couple and condense. The superconducting state is achieved through a Bose-type condensation in a single quantum state with a well-defined global phase, where the superconducting gap Δ_k is determined by the self-consistent equation [12]:

$$\Delta_k(E_k) = \Delta_{0,k} - 2\beta_k P_0(E_k) \quad (2)$$

with $E_k = \sqrt{\epsilon_k^2 + \Delta_k^2}$, the quasiparticle dispersion. The self-consistent solution of this equation at $\epsilon_k = 0$ gives the condensation level $\Delta_k = \Delta_p \cos(2\theta)$. In the antinodal direction ($\theta = 0$), the condensation energy is thus

$$\varepsilon_c = \Delta_0 - \Delta_p = 2\beta P_0(\Delta_p)$$

The unconventional dependence of the interaction term $\beta_k P_0(E_k)$ with the quasiparticle energy E_k expresses the interaction of a quasiparticle with pair excited states (see the diagram, Fig. 1).

The coefficient $\beta_k \propto \beta_0 N_{oc}(T)$ is proportional to the number of pairs in the condensate $N_{oc}(T)$ and plays the role of an order parameter vanishing at T_c . Taking the d -wave symmetry of the SC wave function into account, $N_{oc}(T)$ is given by [12]:

$$N_{oc}(T) = n_0 - \mathcal{A}(T) \int_0^{2\pi} d\theta \int_{\Delta_p \cos(2\theta) + \delta}^{\infty} d\Delta_i$$

$$\times P_0(\Delta_i) f_B(\Delta_i - \Delta_p \cos(2\theta), T) \quad (3)$$

where $f_B(E, T) = (e^{\frac{E-\mu}{k_B T}} - 1)^{-1}$ is the Bose statistics, Δ_p the ground state energy and $\mathcal{A}(T)$ is for normalization. The ‘minigap’ δ separates the condensate from the first excited state and is thus identified as the energy needed to extract a pair from the condensate. As we shall examine below, this quasi-Bose transition is quite distinct from the atomic situation, due to coexisting quasiparticle and normal electron excitations of the condensate.

The interaction term $\beta_k P_0(E_k)$ in the gap equation 2 is responsible for the ‘peak-dip-hump’ structure in the QP excitation spectrum. The fit of the QP DOS using equation 2 allows to extract the SC parameters (Δ_0 , σ_0 , Δ_p , β) with high precision as a function of doping, i.e. the phase diagram. As shown in [12], at $T = 0$, coherence is not revealed in the spectral gap Δ_p but in the fine structure (‘peak-dip-hump’) developing at higher energy.

In this work, we extend the notion of quasiparticle DOS usually associated with the condensate to include the excited states as well. In particular, we calculate the temperature dependence of the QP DOS from the low- T condensed state through the PG state and up to T^* , for a range of doping values from underdoped to overdoped regimes. We show that the contribution of excited states gives rise to a pseudogap at T_c , in full agreement with experiments [9]. In addition, a similar result is found in the vortex core, where coherence is broken by the magnetic field. The coexistence of boson and fermion excitations and their relative contribution to the DOS are the main points of the present work. Our model thus reproduces the temperature dependence of the experimental STM spectra up to T^* (see [9, 10]). It gives direct insight into the new condensate density and the excited-pair density which are original to the model.

Quasiparticle excitation spectrum. –

Case of conventional superconductors. The excitations of the system are important for all thermodynamic and transport properties. In the BCS theory [17], at finite temperature, the presence of condensed Cooper pairs below T_c is accompanied by quasiparticle excitations having the dispersion $E_k = \sqrt{\epsilon_k^2 + \Delta_k(T)^2}$ where ϵ_k is the kinetic energy relative to the Fermi energy and $\Delta(T)$ is the SC gap. The latter decreases as a function of temperature as a result of QP excitations governed by Fermi-Dirac statistics $f(E, T) = (e^{\frac{E-\mu}{k_B T}} + 1)^{-1}$ and eventually vanishes at the critical temperature T_c .

The zero-temperature gap is related to the fundamental parameters and energy scales of the material (Debye frequency, electron-phonon coupling, DOS at the Fermi energy). Proportional to the critical temperature, it has a universal value : $\Delta(T = 0) \approx 1.7 k_B T_c$.

The tunneling density of states (DOS) is a direct consequence of the QP dispersion:

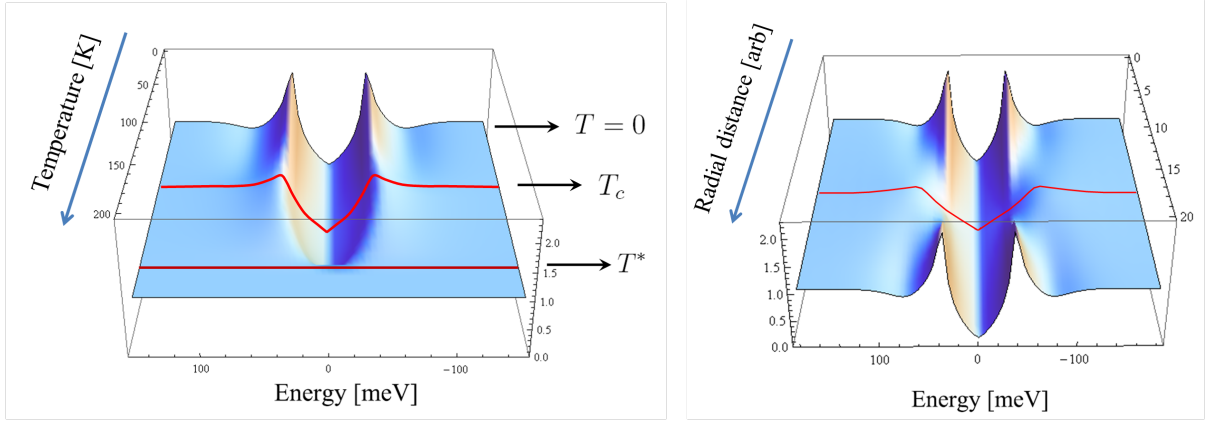


Fig. 2: Left panel: temperature evolution of the QP-DOS from zero temperature through T_c (SC to PG transition) up to T^* (normal state). Right panel: expected evolution of the QP-DOS crossing a vortex core. The coherence peaks disappear leaving a peakless PG spectrum. Note that the additional thermal broadening ($\sim 3.5 k_B T_c$) due to the tunneling process is not included throughout this work.

$$\mathcal{N}_S(E) = \mathcal{N}_n(E_F) \frac{|E|}{\sqrt{E^2 - \Delta(T)^2}} \Theta(|E| - \Delta) \quad (4)$$

where $\mathcal{N}_n(E_F)$ is the normal DOS at the Fermi energy and $\Theta(z)$ the heaviside function.

At low temperatures, it reveals a sharp energy gap at the Fermi-level - first measured by I. Giaever using planar junctions [18] - delimited by high symmetric peaks. The DOS has since been measured many times with a high precision by tunneling spectroscopy using planar junctions or STM geometries [19] and later on using Angle Resolved Photon Emission Spectroscopy (ARPES) in cuprates [20, 21].

Unconventional superconductors. In our model, the situation is radically different from the BCS situation. In this case, the gap does not vanish at T_c but at T^* , the pair formation onset temperature. On the other hand, the condensation energy $\varepsilon_c \propto \beta_0 N_{oc}(T)$ vanishes at T_c , which is much smaller than T^* . Thus, for the temperature dependent DOS, two additional contributions arise, absent in the BCS case: first the contribution of excited pairs increasing with temperature up to T_c , and second the dissociation of pairs increasing up to T^* .

The DOS as a function of temperature for optimal doping is reported in Fig. 2, left panel. At zero temperature, it exhibits the characteristic shape of the excitation spectrum of the cuprates, with the characteristic ‘peak-dip’ features. As the temperature rises, the quasiparticle peak heights decrease while the *gap width* remains roughly constant, contrary to the BCS case. At T_c , there is a clear pseudogap with attenuated QP peaks. For higher temperature ($T > T_c$), one notes the filling of states inside the gap while the QP peaks are further smoothed. The gap eventually disappears completely at T^* .

Note that other theories have been proposed for the ‘peak-dip-hump’ structure in the DOS measured by

STM/ARPES. The most popular approach is the coupling to a spin collective mode (see [22], [23] and reference therein). It appears that the fit to the experimental spectra is rendered difficult in this framework due to the necessarily retarded interaction. Furthermore, to our knowledge, its extension to finite temperature has not been done.

To the contrary, the pair-pair interaction model accurately reproduces the DOS measured by STM/STS in $\text{BiSr}_2\text{Ca}_2\text{Cu}_2\text{O}_{8+\delta}$ [9] or $\text{TlBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8.5+\delta}$ [10]. As observed in experiments [7, 24, 25], we find a similar situation as function of the magnetic field, crossing a vortex (see Fig. 2, right panel). Here we assume that the effect of magnetic field is to reduce the coherence energy β . Quite analogous to the effect found in case of a disorder potential [3, 26]. The coherence peaks decrease when approaching the vortex center where finally a peakless pseudogap is found in the vortex core. In a general way, the loss of coherence of the SC to PG transition is driven by the vanishing of β , the pair-pair interaction.

DOS in the pair-pair interaction model. – Contrary to standard models, we express the quasiparticle DOS as a sum of three terms:

$$\mathcal{N}(E, T) = \mathcal{N}_{cond}(E, T) + \mathcal{N}_{ex}(E, T) + \mathcal{N}_{diss}(E, T) \quad (5)$$

where the distinct contributions are:

- (i) the condensate, $\mathcal{N}_{cond}(E, T)$
- (ii) the excited pairs, $\mathcal{N}_{ex}(E, T)$
- (iii) the dissociated pairs, $\mathcal{N}_{diss}(E, T)$

Condensate. The contribution of the condensate $\mathcal{N}_{cond}(E, T)$ is proportional to the number of pairs in the condensate at temperature T :

$$\mathcal{N}_{cond}(E, T) = N_{oc}(T) \times \mathcal{N}_{SC}^d(E, T) \quad (6)$$

where $\mathcal{N}_{SC}^d(E, T)$ is the zero-temperature superconducting DOS, found when all pairs belong to the condensate, i.e. when $N_{oc}(T=0) = n_0$.

As shown in Ref. [12, 27], neglecting any QP lifetime broadening, the condensate DOS along the θ direction is:

$$\begin{aligned}\mathcal{N}_{SC}^d(E, \theta) &= \frac{\mathcal{N}_n(E_F)}{2\pi} \int_0^\infty d\epsilon_k \delta(E_k - E) \\ &= \frac{\mathcal{N}_n(E_F)}{2\pi} \left[\frac{\partial \epsilon_k}{\partial E_k} \right]_{E_k=E}\end{aligned}\quad (7)$$

with

$$\frac{\partial \epsilon_k}{\partial E_k} = \frac{E_k - \Delta_k(E_k, \theta) \frac{\partial \Delta_k}{\partial E_k}}{\epsilon_k} = \frac{E_k - \Delta_k(E_k, \theta) \frac{\partial \Delta_k}{\partial E_k}}{\sqrt{E_k^2 - \Delta_k(E_k)^2}}$$

where $\mathcal{N}_n(E_F)$ is the normal DOS at the Fermi energy. The superconducting DOS is thus inversely proportional to the slope of $E_k(\epsilon_k)$, depending explicitly on the gap function (2). Therefore, the fine structure in the DOS, the ‘peak-dip-hump’ features, originates from the interaction term, $2\beta_k P_0(E_k)$, of the gap equation. While these features have been studied at $T=0$ [12, 27], in the following we address the question of their temperature dependence.

Excited pairs. As soon as the temperature increases, excited pairs start to contribute (see Fig. 3) to the DOS. This leads to the term:

$$\mathcal{N}_{ex}(E, T) = \mathcal{A}(T) \sum_{\epsilon_i > \delta \cos(2\theta)} f_B(\epsilon_i, T) \mathcal{N}_i(E, \Delta_i) \quad (8)$$

where $\epsilon_i = \Delta_i - \Delta_p \cos(2\theta)$ is the excitation energy and $f_B(E, T)$ is the Bose distribution. $\mathcal{N}_i(E, \Delta_i)$ is the DOS associated with each pair Δ_i . In the standard d -wave form, assuming that excited pairs are not interacting, we have:

$$\mathcal{N}_i(E, \Delta_i) = \frac{\mathcal{N}_n(E_F)}{2\pi} \int_0^{2\pi} d\theta \frac{|E|}{\sqrt{E^2 - (\Delta_i \cos(2\theta))^2}}$$

Dissociated pairs. The bosons in our model are composite fermions but, distinct from the BCS case, as the temperature rises an increasing fraction of the pairs *dissociate at the Fermi energy*. This appears as some of the Δ_i vanish, i.e. when $\Delta_p(T)$ starts to decrease. This gives rise to the third term:

$$\mathcal{N}_{diss}(E, T) = \mathcal{N}_n(E_F) \times N_{diss}(T) \quad (9)$$

Here $N_{diss}(T)$ is the number of dissociated pairs at the temperature T (see Fig. 3). It increases up to T^* , where finally $N_{diss}(T^*) = n_0$.

Discussion. –

Pair densities. The number of pairs (including dissociated pairs) must follow a sum rule:

$$N_{oc}(T) + N_{ex}(T) + N_{diss}(T) = n_0 \quad (10)$$

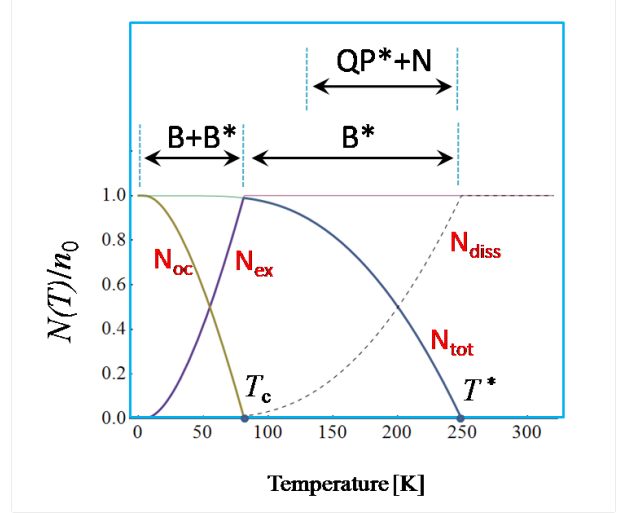


Fig. 3: Plot of the 4 important occupation densities (underdoped case): n_0 the total number of pairs at zero temperature, N_{oc} the number of pairs in the condensate, N_{ex} the excited bosons, N_{diss} the dissociated pairs. The upper part of the figure shows the characteristic temperature ranges in the model: B – boson of the condensate (vanishing at T_c), B^* – excited boson (maximum at T_c but vanishing at T^*), QP^* – quasiparticles excitations (also vanishing at T^*), N – ‘normal’ electrons arising from pair dissociation (maximum at T^*).

This rule involving the number of pairs guarantees the conservation of states in the DOS at any temperature T :

$$\int_{-\infty}^{\infty} dE \mathcal{N}(E, T) = C$$

where C is a constant. In addition, we assume that the total number of pairs (i.e. condensed plus excited pairs) must follow

$$N_{tot}(T) = N_{oc}(T) + N_{ex}(T) \sim \Delta_p(T)^2$$

which vanishes as $\sim |T - T^*|$ near T^* . Note that the precise shape of $N_{tot}(T)$ has never been investigated. Our assumption on its particular temperature dependence does not influence the basic conclusions of the paper.

As previously noted, at $T=0$, all pairs belong to the condensate: $N_{oc}(T=0) = n_0$. As the temperature increases, pairs are progressively excited from the condensate, i.e. $N_{ex}(T)$ increases with temperature in opposition to $N_{oc}(T)$, as in Fig. 3. Only low-lying excited states dominate owing to the boson occupation $f_B(E, T)$ in equation (3). With temperature, the condensate empties while the number of excited pairs increase. Finally, at the critical temperature, there is no longer a condensate:

$$N_{oc}(T_c) = 0$$

In the underdoped regime, where $T_c \ll T^*$ (the case of Fig. 3), the number of dissociated pairs is negligible, so that all pairs are excited at T_c :

$$N_{ex}(T_c) \approx n_0$$

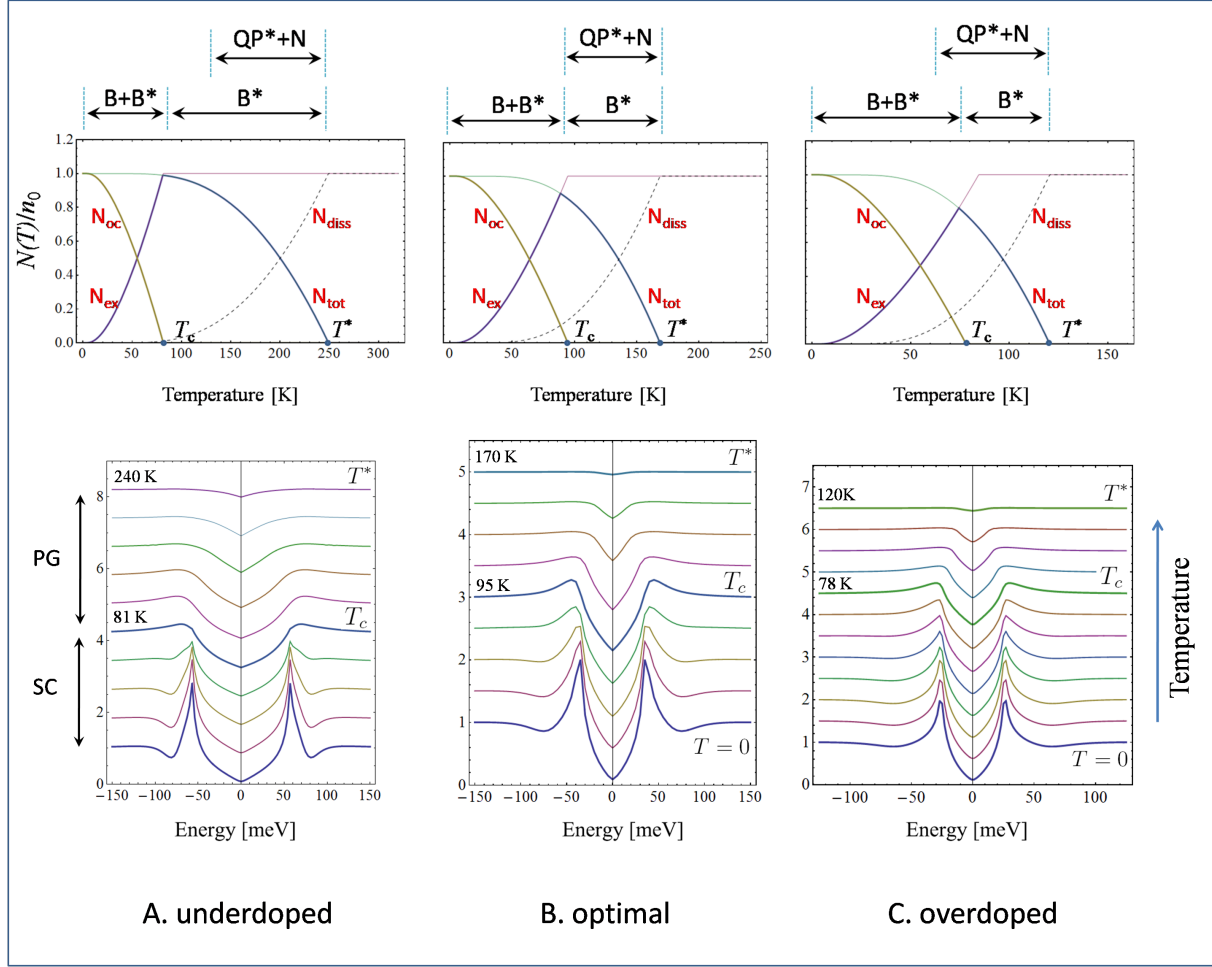


Fig. 4: Comparison of the T -dependent DOS for three different doping levels: underdoped (left panel), optimal (middle panel), overdoped (right panel). The corresponding temperatures (T_c , T^*) are indicated. The upper figures show the temperature range of the boson and fermion excitations. It is important to note the striking difference between the onset of quasiparticle and pair dissociation, for the overdoped and underdoped cases.

In the more general case, to be discussed below, some pairs are already dissociated at the critical temperature and one has:

$$N_{diss}(T_c) = n_0 - N_{ex}(T_c) > 0$$

The effect of pair dissociation is highly sensitive to the $(T^* - T_c)$ difference, in practice to the doping value. An in-depth work of Yazdani et al. [28–30] investigated the zero bias conductance in the SC and PG states. While it is quite uniform in the superconducting state, it showed significant variations above T_c . This result can be directly explained by the presence of dissociated pairs, giving rise to the third term in equation (5).

It is important to stress that the progressive broadening of the QP peaks from SC to PG state is not due to a finite quasiparticle lifetime (Γ_{Dynes} broadening [31]). Instead, it results from the second term in equation 5 due to excited pairs existing at finite temperature. The calculation leads to a larger PG in the vortex core (see Fig. 2, right panel), where it directly stems from the distribution of preformed pairs $P_0(\Delta_i)$, rather than at T_c where it also

involves the Bose statistics $f_B(E, T)$. Such a filling of the DOS is a direct consequence of the statistical occupation of pair excitations inherent to the model.

Changes of the DOS with doping. Using the previous relations, we have calculated the temperature evolution of the DOS for three different doping levels corresponding to the underdoped regime (Fig. 4A), optimally doped (Fig. 4B) and overdoped regime (Fig. 4C). At low temperature, the excitation spectrum exhibits the characteristic d -wave shape of HTc cuprates (see [6] for a review) and the ‘peak-dip’ features. Note that it is more pronounced in the underdoped case, while it is slightly attenuated in the overdoped case.

The coherence peaks broaden as the temperature rises, as a consequence of the increasing contribution of incoherent excited pairs, and eventually vanish at T_c , where all bosons have left the condensate. The contribution of $N_{ex}(T)$ gives rise to a clear pseudogap at T_c for the three different doping values.

The remarkable difference between underdoped and

overdoped samples emerges as the limit at which quasiparticle QP* and normal electrons appear (it is defined as the temperature at which the total number of pairs $N_{tot}(T)$ decreases significantly, i.e. when it reaches $\sim 0.9 n_0$). They originate respectively from excited pairs Δ_i , which give rise to quasiparticles $E_k^i = \sqrt{\epsilon_k^2 + \Delta_i^2}$, and from dissociated pairs (with energy $\sim 2\epsilon_k$). The presence of normal electrons implies that some states are present inside the gap, leading to a finite value in the DOS at $E = 0$. As a rule of thumb, pair excitations attenuate the QP peaks while the normal states fill in the gap.

The situation is quite extreme in the underdoped case, wherein T_c is much smaller than T^* (Fig. 4A, upper panel). Consequently quasiparticles QP* and normal electrons arise well above T_c . In this case, the SC to PG transition is dominated by boson excitations, the fermions contribution being negligible. This quasi-Bose transition is characterized by the vanishing of the condensate (B) at T_c leaving only pair excitations (B*) just above T_c . The opposite scenario is found in the overdoped regime where quasiparticles and normal electrons coexist *below* T_c . Finally, in the optimally doped regime, with the highest T_c , it is remarkable that the onset of quasiparticle and normal excitations (QP*+N, in Fig. 4B) coincides with the critical temperature.

Given their importance, further experiments should be done to verify the density profiles with the predictions of the model.

Conclusion. – In summary, we proposed a comprehensive model for HTc superconductors based on an underlying disordered state of preformed pairs. Coherence is achieved by means of the pair-pair interactions leading to a Bose-type condensation. Contrary to the standard BCS theory, several ingredients contribute to the DOS at finite temperature: i) the condensate, ii) the presence of excited pairs, iii) normal electrons arising from pair dissociation.

As a direct consequence of our model, the first contribution decreases with temperature, while the two other contributions, preformed pairs and normal electrons, coexist at finite temperature. The low temperature DOS exhibits the well-known ‘peak-dip’ structure - directly related to the pair-pair interaction - which characterizes superconducting coherence. Due to the presence of excited pairs, the quasiparticle peaks in the DOS decrease progressively, concomitant with the pair-pair interaction, as a function of temperature and a pseudogap is finally found at T_c up to T^* in the whole doping range. A similar pseudogap is found in the vortex core, in very good agreement with tunneling experiments.

The evolution of the DOS as a function of temperature reveals a mixed state wherein condensed or excited bosons and normal electrons coexist.

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